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Title: NOX REMOVAL IN JET-ENGINE TEST CELL EXHAUST:
PROPOSED NON-THERMAL PLASMA SYSTEMS AND
ECONOMIC CONSIDERATIONS

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NO_x Removal in Jet-Engine Test Cell Exhaust: Proposed Non-Thermal Plasma Systems and Economic Considerations

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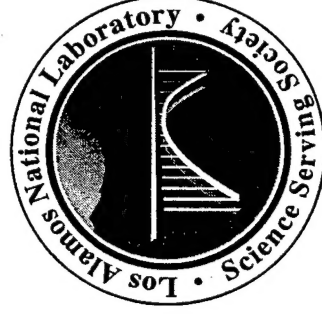J.-S. Chang, K. Urashima, and S.J. Kim
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Thanks to

the US Strategic Environmental Research & Development Program (SERDP)

for support for this work.

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Outline of talk

- Motivation for and applications of research

- Background of atmospheric-pressure, non-thermal plasma processing for environmental applications (air-pollution control)

  ◆ Basics of radical formation & decomposition chemistry

  ◆ Figures-of-merit & costs for decomposition of example pollutants

- Removal of NO_x and VOCs from jet-engine test facilities

  ◆ Plans for field demonstration of pilot unit

  ◆ Candidate NTP systems

  ◆ Economic considerations (candidate systems compared to conventional)

- Future trends

- Summary

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Technology-development motivators are regulatory, economic, and social

Regulatory
- New and/or more stringent regulations
  Clean Air Act Amendment of 1990, Clean Water Act
  Attendant MACT standards

- Enforcement of regulations

Economic
- Impact of regulations
- Costs associated with conventional methods

Social
- Environmental consciousness
- Health risks

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The CAAA 1990 contains three titles closely related to NTPs

Title III (Air Toxics)
- Directly concerned with emission of hazardous air pollutants (HAPs)
- Established standards for 189 listed chemicals

Title IV (Acid Rain)
- Reduction of SOx/NOx emissions

Title VI (Stratospheric Ozone)
- Phase out of VOCs

MACT (Max Achievable Control Technology) Standards
- Calls for state-of-art pollution control equipment
- Require changes in equipment, processes, treatment

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Regulations for engine test facilities are changing under CAAA 1990 10-yr provisions

- Previously, engine test facilities were considered mobile sources because engines were moved in and out of the facilities for testing operations. However, the test facility itself was usually a permanent structure.

- The upcoming new standards will consider the test facilities as stationary sources, which are regulated differently (more stringently) than mobile sources.

- The test-facility emissions (primarily NO_x and volatile organic compounds - VOCs) have dilute concentrations (e.g., < 50 ppm) and very high ($\sim 10^6 - 10^7$ Nm^3/h) exhaust-gas flows.

- Consequently, new emissions-control technologies are being explored.

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Non-thermal plasmas (NTPs) are a type of advanced oxidation & reduction process which use free radicals to decompose pollutants in the gas phase.

Non-thermal plasmas are a type of AOP making use of "cold combustion" via free-radical reactions.

Electrons are energetic (*hot*), while neutral & ions are at near-ambient temperatures (*cold*).

The key idea is to direct electrical energy into favorable chemistry for oxidizing and/or reducing pollutants to more manageable forms (simpler or mineralized terminal products).

Applications

- **Flue gases & engine emissions:** e.g., NO_x & SO_x
- **VOCs:** e.g., hydrocarbons & halocarbons
- **Odors:** H_2S, others

Potential Advantages

- In-situ generation of chemical reactants
- No added fuel (greenhouse gases)
- Simultaneous removal of multiple pollutant species
- Electronic feedback for optimal process control.

Non-thermal Plasmas Decompose Pollutants Via Active Species Generated in the Process Gas

Electrical Energy

Effluent Stream
(w/decomposed pollutants, simpler compounds)

Plasma & active species
(e.g., free radicals)

Influent Stream
(w/pollutants)
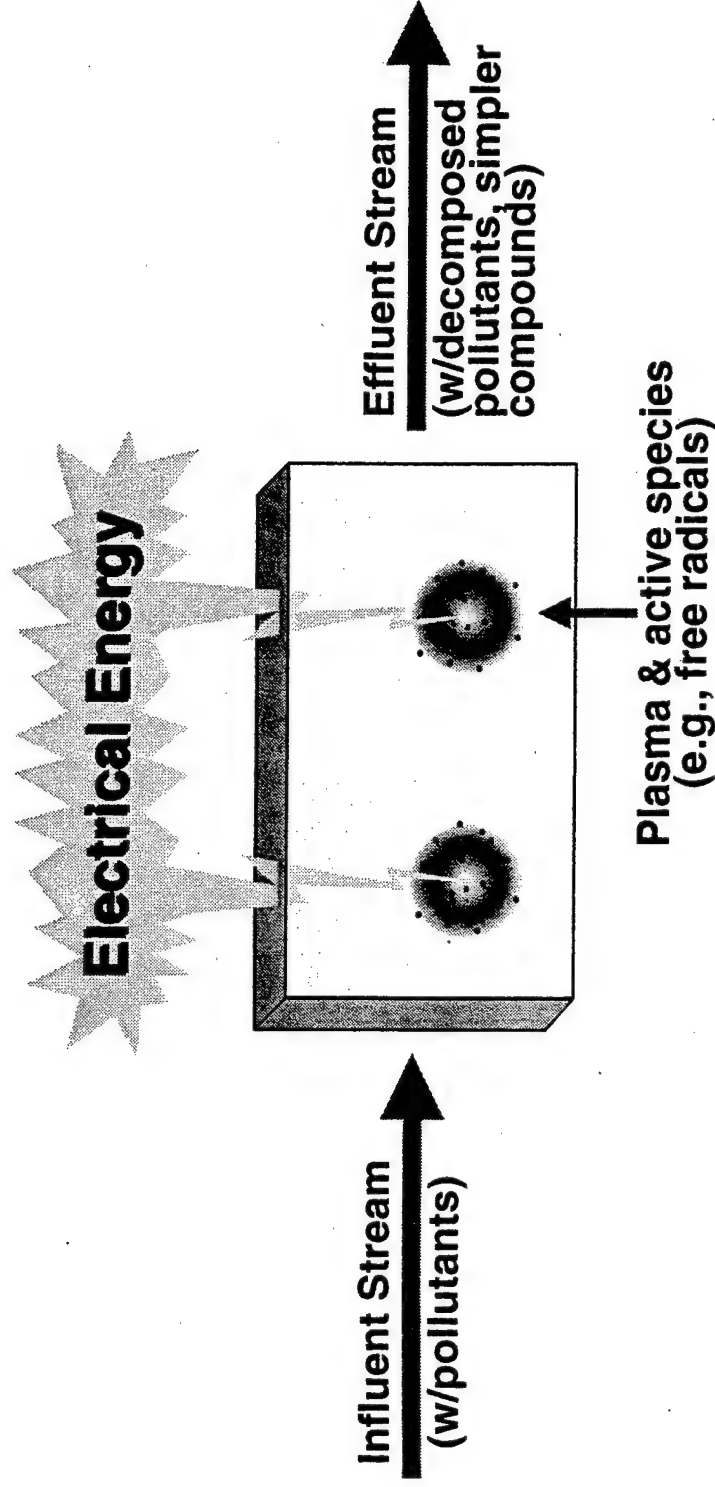
The radical production efficiency (G-value) depends on the gaseous electronics/plasma chemistry.

$$G = f\left(\frac{k_{rad}}{\mathbf{V}_d \frac{E}{N}}\right)$$

- Radical generation is mainly initiated by energetic-electron collisions,

- E/N is the reduced field,

- $\mathbf{V}_d$ is the electron drift velocity, which depends on E/N,

- k_{rad} is the rate constant for radical formation (e.g., a dissociation rate constant, which depends on E/N), and/or other rate constants.

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The overall process efficiency can be effectively separated into two terms: the radical production efficiency and the radical utilization efficiency.

$$\eta_{net} = G_R \, \eta_u$$

- G_R is the radical-production efficiency (mainly depends on gaseous electronics/plasma chemistry)

- η_u is the radical utilization efficiency (mainly depends on chemical kinetics)

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Gas-phase plasma chemical decomposition is driven by electron impact and radical attack.

(1) $e + X \rightarrow$ products

(2) $O, OH, N,$ etc. $+ X \rightarrow$ products

- The first reaction is dominant at large pollutant mole fractions.

- The second dominates at smaller mole fractions.

(More energy is directly absorbed by pollutant at high mole fraction, hence electron channel dominates.)

Plasma chemical decomposition of VOCs produces a variety of terminal products.

$e, O, OH, N,$ etc. $+ X \rightarrow$ **products**

- **Manageable products:** CO_2, CO, Cl_2, HCl, $COCl_2$

- **Undesirable products:** Other halocarbons, hazardous byproducts (e.g., DCAC – $CHCl_2COCl$); polymers

- **Secondary treatment:**
  $CO_2 + NaOH \rightarrow NaHCO_3$
  $CO + catalyst \rightarrow CO_2$
  $Cl_2 + NaHCO_3 \rightarrow NaCl + HCl + \dots$
  $HCl + NaOH \rightarrow NaCl + H_2O$
  $COCl_2 + H_2O \rightarrow 2HCl + CO_2$

Major de-NO$_x$ Reactions in Moist Gas Mixtures w/o HCs

$O + NO + M \rightarrow NO_2 + M$

$O + NO_2 \rightarrow NO + O_2$

$O + N_2 \rightarrow NO + N$

$O + O_2 + M \rightarrow O_3 + M$

$O_3 + NO \rightarrow NO_2 + O_2$

$O_3 + NO_2 \rightarrow NO_3 + O_2$

$NO_2 + NO_3 + M \rightarrow N_2O_5 + M$

$N(^2D) + O_2 \rightarrow NO + O$

$N + NO \rightarrow N_2 + O$

$N(^2D) + NO \rightarrow N_2 + O(^1D)$

$N + NO_2 \rightarrow N_2O + O$

$N + NO_2 \rightarrow 2NO$

$N(^2D) + H_2O \rightarrow NH + OH$

$NH + NO \rightarrow N_2 + OH$

$NH + O_2 \rightarrow NO + OH$

$NO + NO_3 \rightarrow 2NO_2$

$NO + OH + M \rightarrow HNO_2 + M$

$OH + HNO_2 \rightarrow NO_2 + H_2O$

$OH + NO_2 + M \rightarrow HNO_3 + M$

$N + NO_2 \rightarrow N_2 + O_2$

$N + N_2O \rightarrow NO + N_2$

$O(^1D) + NO_2 \rightarrow NO + O_2$

$O(^1D) + N_2O \rightarrow 2NO \text{ or } N_2 + O_2$

$N_2(A) + O_2 \rightarrow N_2 + 2O$

$N_2(A) + N_2O \rightarrow N_2 + N + NO$
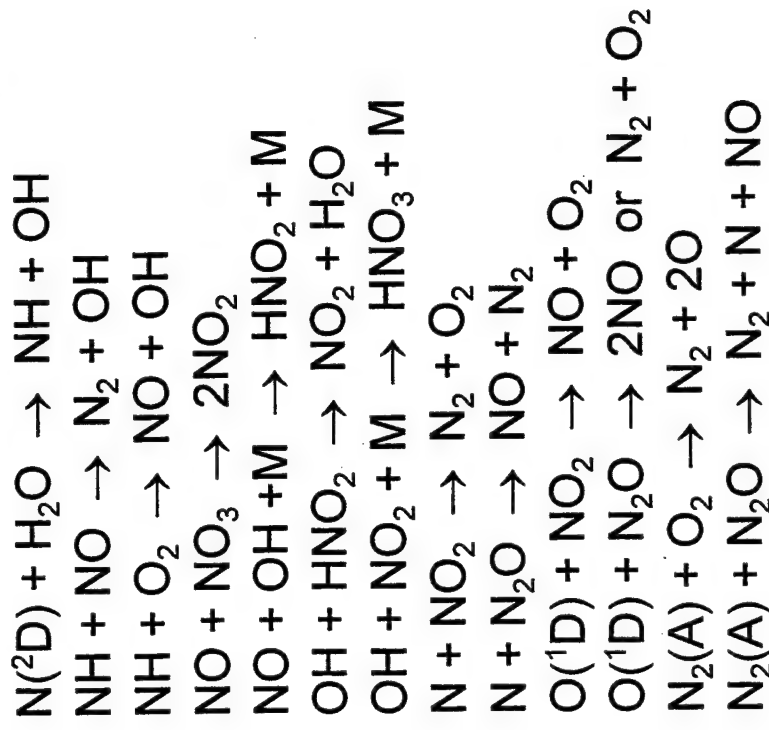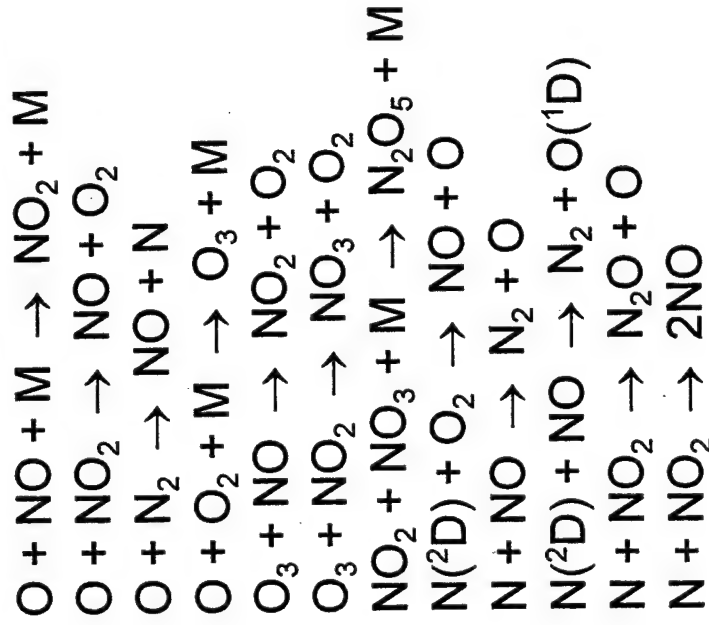
Primary Acid-Formation Pathways:

$NO + OH + M \rightarrow HNO_2 + M$

$OH + NO_2 + M \rightarrow HNO_3 + M$

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The presence of SO_2 recycles OH radicals & reduces effective de-NO_x energy cost

$OH + SO_2 + M \rightarrow HSO_3 + M$

$HSO_3 + O_2 \rightarrow HO_2 + SO_3$

$HO_2 + NO \rightarrow NO_2 + OH$

The OH radical then goes on to be used again in de-NO_x

$OH + NO + M \rightarrow HNO_2 + M$

$OH + HNO_2 \rightarrow NO_2 + H_2O$

$OH + NO_2 + M \rightarrow HNO_3 + M$
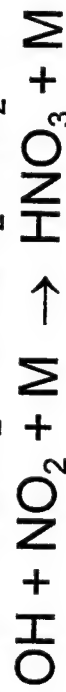
Acid is also formed by the reaction

$HSO_3 + H_2O \rightarrow H_2SO_4$

With ammonia (NH_3) addition, useful particulates (fertilizer) can be formed from NO_x

$e + NH_3 \;\rightarrow\; NH_2 + H + e$

$e + NH_2 \;\rightarrow\; NH + H + e$

$NH + H \;\rightarrow\; N + H_2$

$NH_2 + NO \;\rightarrow\; N_2 + H_2O$

$NH + NO \;\rightarrow\; N_2 + OH$

$N + NO \;\rightarrow\; N_2 + O$

$OH + NH_3 \;\rightarrow\; NH_2 + H_2O$

$OH + NO + M \;\rightarrow\; HNO_2 + M$

$OH + HNO_2 \;\rightarrow\; NO_2 + H_2O$

$OH + NO_2 + M \;\rightarrow\; HNO_3 + M$

$NH_3 + HNO_3 \;\rightarrow\; NH_4NO_3$ (Ammonium Nitrate fertilizer)

A generalized removal equation depends on plasma chemistry (radical yields), reaction chemistry, and applied plasma specific energy.

Generalized differential equation:

$$\frac{k[X] + \sum_i k_{S_i}\,[S_i]}{k[X]}\,d[X] = -\,G\,d\overline{E}$$

Integration with limits $[X]_0 \to [X]$ and $0 \to E$ gives:

$$\frac{[X]}{[X]_0} + \frac{\sum_i k_{S_i}\,[S_i]}{k\,[X]_0}\,\ln\frac{[X]}{[X]_0} - 1 = -\frac{G\overline{E}}{[X]_0}$$

For low degree of removal (i.e., $[X]/[X]_0 \sim 1 + \ln\{[X]/[X]_0\}$), an analytical solution is obtained:

$$[X] / [X]_0 = \exp\left(-\overline{E}/\beta\right)$$

where

$$\beta = \frac{1}{G}\left([X]_0 + \frac{\sum_i k_{S_i}\,[S_i]}{k}\right)$$

When $k[X] \ll \sum_i k_{S_i}\,[S_i]$, the β-value and, hence the degree of removal $[X]/[X]_0$ shows no dependence on the initial concentration $[X]_0$.

Example scaling laws for pollutant removal (non-sensitive $[X]_0$ dependence)

(1) $[X] = [X]_0 \exp(-\bar{E}/\beta)$,

where $[X]_0$ is the initial pollutant concentration, $[X]$ is the resulting concentration, $\bar{E}$ is the applied specific energy (or plasma power divided by gas flow rate, P/Q), and β is the e-fold energy density. Supplying one β to the reactor reduces the concentration by 1/e, 2β by $1/e^2$, and so on.

A useful figure of merit for the decomposition of pollutants is defined by the energy delivered to the plasma per hazardous molecule removed from the gas stream. At any instant, this can be expressed as the following quantity obtained by solving Equation 1 for $\bar{E}$ and taking the derivative:

(2) $\gamma_i = -\dfrac{d\bar{E}}{d[X]} = -\dfrac{d[-\beta \ln(\dfrac{[X]}{[X]_0})]}{d[X]} = \dfrac{\beta}{[X]}$.

This is the instantaneous energy cost per molecule removed.

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Scaling laws for pollutant removal (cont'd)

A more practically-useful parameter is the integral, or average, energy cost γ

$$(3) \quad \gamma = \frac{\bar{E}}{[X]_0 - [X]} = \frac{-\beta \ln(\frac{[X]}{[X]_0})}{[X]_0(1 - \frac{[X]}{[X]_0})} \qquad (= \beta/[X]_0 \text{ (at optimum; i.e., } [X]/[X]_0 \sim 1).$$

Here, the energy cost per molecule is expressed in terms of the β-value, the degree of removal, and the initial concentration.

When radical-pollutant attack dominates $\beta \approx [X]_0/G$ and the removal cost is then independent of the initial concentration

$$(4) \quad \gamma = \frac{\bar{E}}{[X]_0 - [X]} = \frac{-\ln(\frac{[X]}{[X]_0})}{G(1 - \frac{[X]}{[X]_0})} \qquad (= 1/G \text{ at optimum; i.e., } [X]/[X]_0 \sim 1).$$

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Scaling laws for pollutant removal (graphical forms)
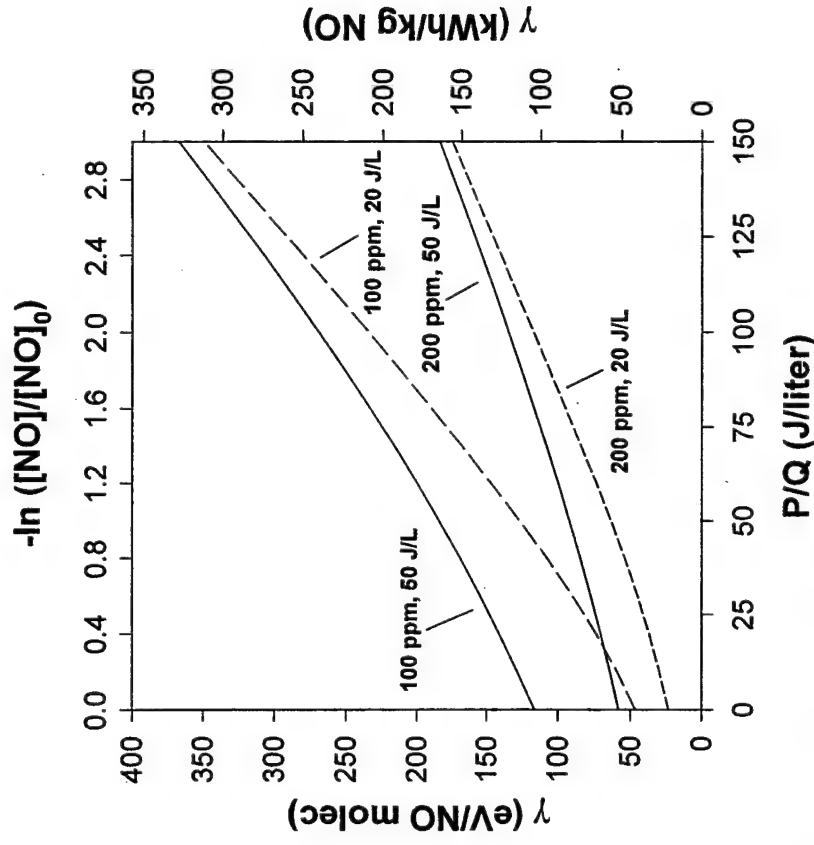
Energy costs and degree of removal for NO removal in an NO-air mixture with one e-fold plasma specific energies of 50 J/liter and 20 J/liter. NO is a compound whose β-value is not very sensitive to the initial concentration; therefore, the removal energy costs decrease as the concentration increases (note 200 ppm case vs 100 ppm case).

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NTP pilot-unit setup for de-NOx field tests

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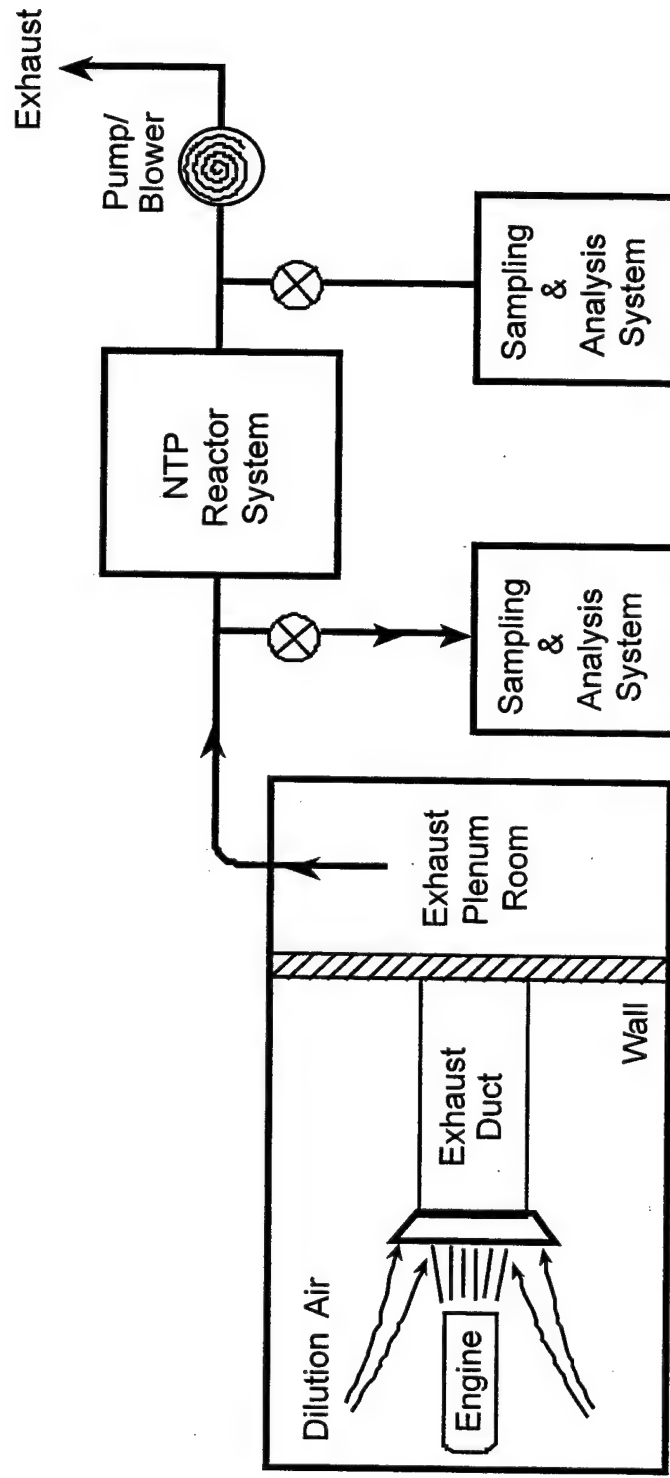
Expected exhaust-gas data and conditions

Source Data	Variable	Units	JETC Values	CMTC Values
Gas Flow	Q_{gas}	Nm^3/h	1.0E+05 1.7E+06	6.6E+03 1.3E+04
Fuel			JP-8	JP-10
Final Exhaust-Gas Composition				
N_2	C_{N2}	%	80.98	78.00
O_2	C_{O2}	%	18.00	21.00
CO_2	C_{CO2}	%	0.50	0.50
H_2O	C_{H2O}	%	0.50	0.50
Density (Normal)	D_{gas}	kg/Nm^3	1.283	1.283
Exhaust Gas Temperature	T_{gas}	C	25	25-75
NTP Inlet Temperature	T_{NTPin}	C	25	25-30
Pressure	Pr_{gas}	mm Hg	720	720
Emission Data				
NO_x	C_{NOx}	ppm	36.00	<10
SO_2	C_{SO2}	ppm	4.59	~1
HC (VOC)	C_{HC}	ppm	60.00	20-25
CO	C_{CO}	ppm	53.36	~5
Particles	C_{part}	mg/Nm^3	-	~/> 1.0
NH_3 Stoichiometric Ratio to NO and SO_2: 1.5 for both JETCs and CMTCs				

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NTP de-NOx System Examples

Stand-Alone System

NO + VOCs + Air → [NTP Reactor] → NO_2, HNO_3, N_2

NO → NO_2 Conversion + Scrubber System

NO + VOCs + Air → [NTP Reactor] → NO_2 HNO_3 → [Wet Scrubber] → Dilute HNO_3 (w / $Na_2S_2O_3$)
$NaNO_3$ + H_2O (w / NaOH)
$Ca(NO_3)_2$ + H_2O (w / $Ca(OH)_2$)

Injection Systems

NO + VOCs + Air → [NTP Reactor] → NH_4NO_3 → [ESP] → Particulate NH_4NO_3 (Fertilizer)
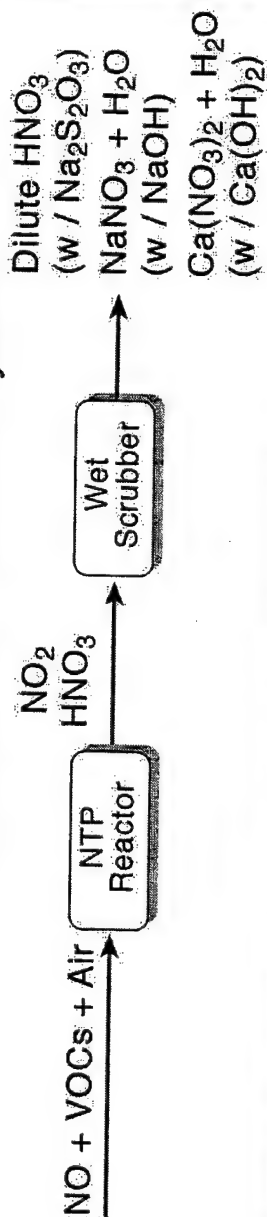
↑ Injection Gas

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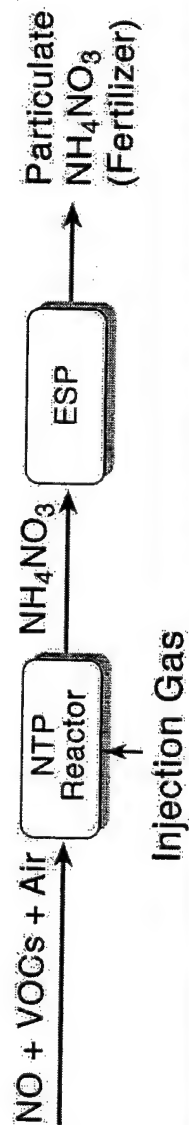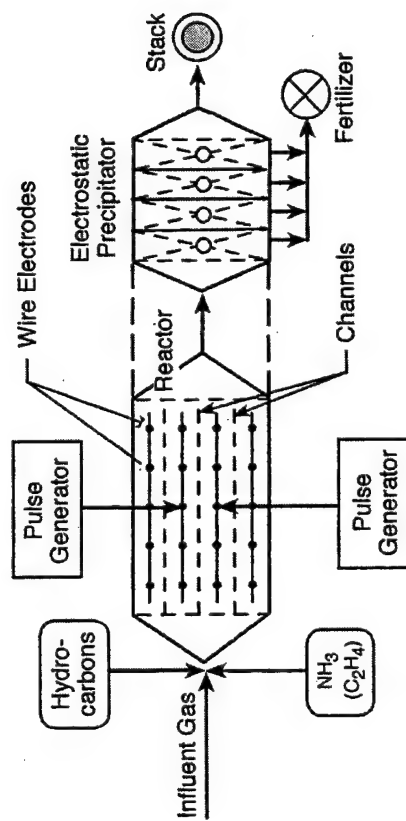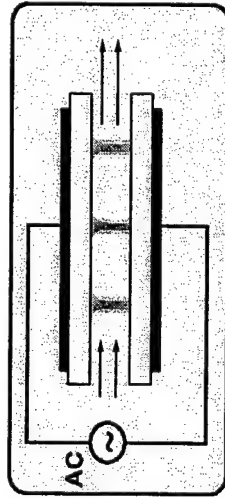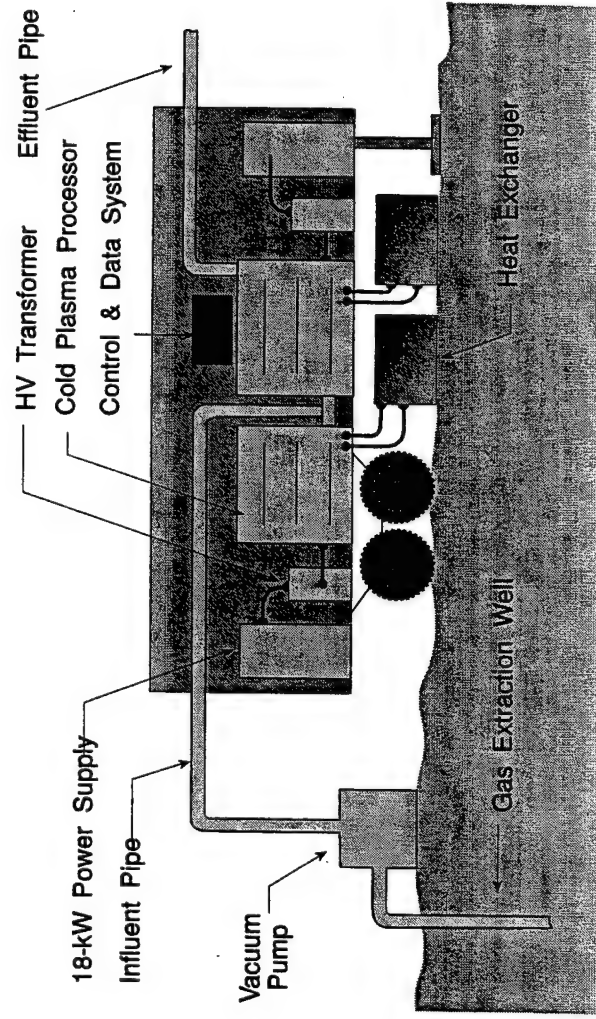
Pulsed Corona System
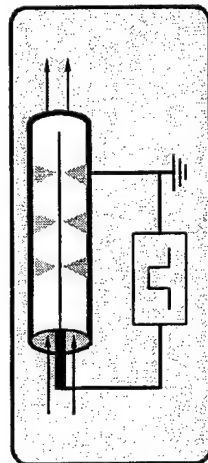
Pulsed or DC corona

Dielectric-Barrier (Silent Discharge) System

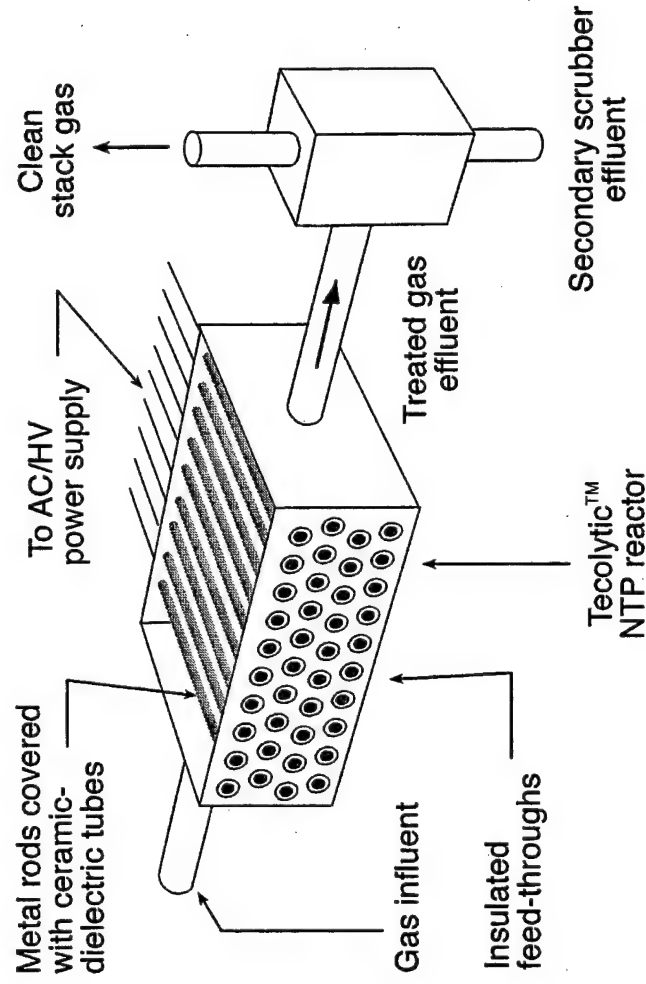Silent discharge
(dielectric-barrier discharge)

Illustration of mobile dielectric-barrier NTP reactor system employed for VOC decomposition tests at McClellan AFB. Each plasma reactor tank operated at up to 10 kW of plasma power.
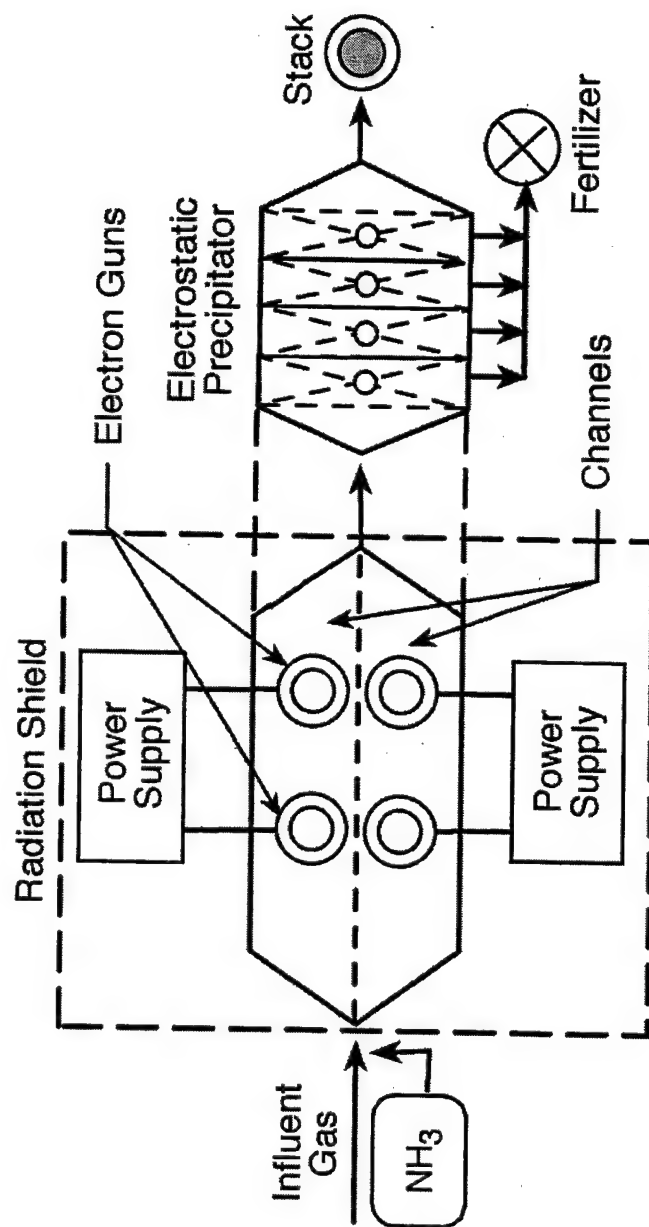
CRADAs with the Electric Power Research Institute (EPRI) & High Mesa Technologies (HMT) were an essential part of the development & fielding of this equipment.

18-kW Power Supply
Influent Pipe
Vacuum Pump
HV Transformer
Cold Plasma Processor
Control & Data System
Effluent Pipe
Heat Exchanger
Gas Extraction Well

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Schematic diagram of commercial Tecolytic™ modified dielectric-barrier NTP reactor system for de-NO$_x$/SO$_x$ (flue-gas treatment)

Clean stack gas

To AC/HV power supply

Treated gas effluent

Secondary scrubber effluent

Metal rods covered with ceramic-dielectric tubes

Tecolytic™ NTP reactor

Gas influent

Insulated feed-throughs

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Electron-Beam System

Stack

Electron Guns

Electrostatic
Precipitator

Fertilizer

Channels

Radiation Shield

Power
Supply

Power
Supply

Influent
Gas

NH₃

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Corona Radical Shower System
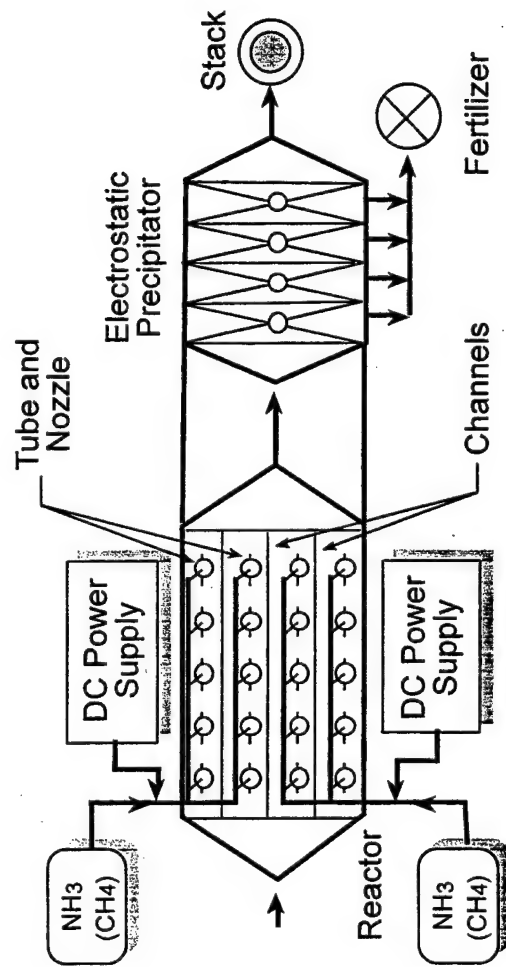
Schematic diagram of CRS reactor. Ammonia (NH_3) or methane (CH_4) are added to generate radicals that drive reactions leading to the formation of particulates; these particulates are then captured by the electrostatic precipitator. Some of the captured products are useful for agricultural fertilizer (e.g., ammonium nitrate, NH_4NO_3).

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Corona Radical Shower (CRS) Lab-Scale Prototype at McMaster University
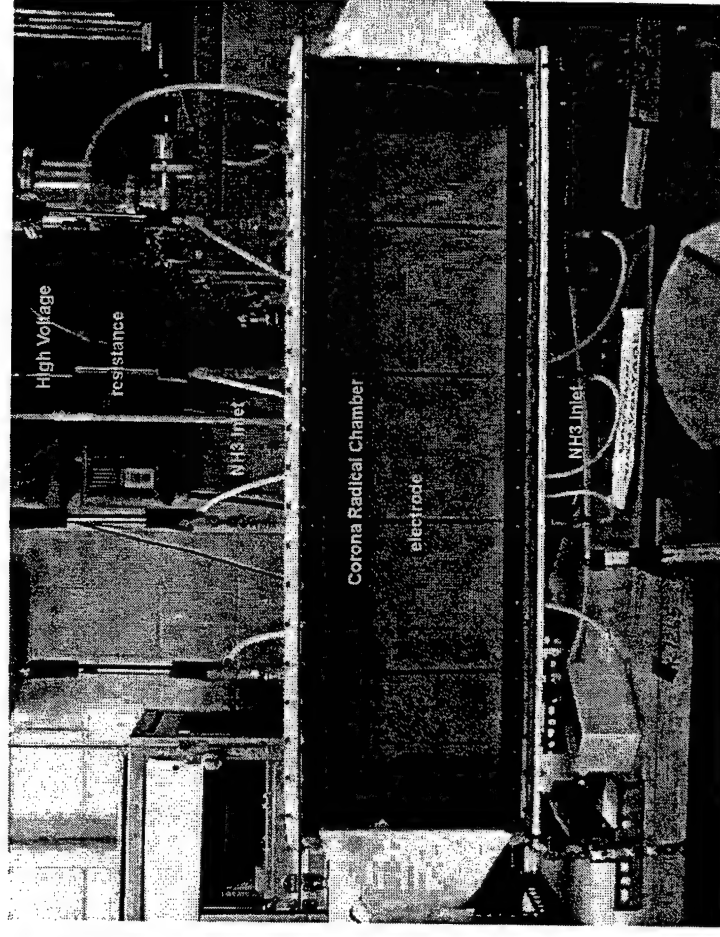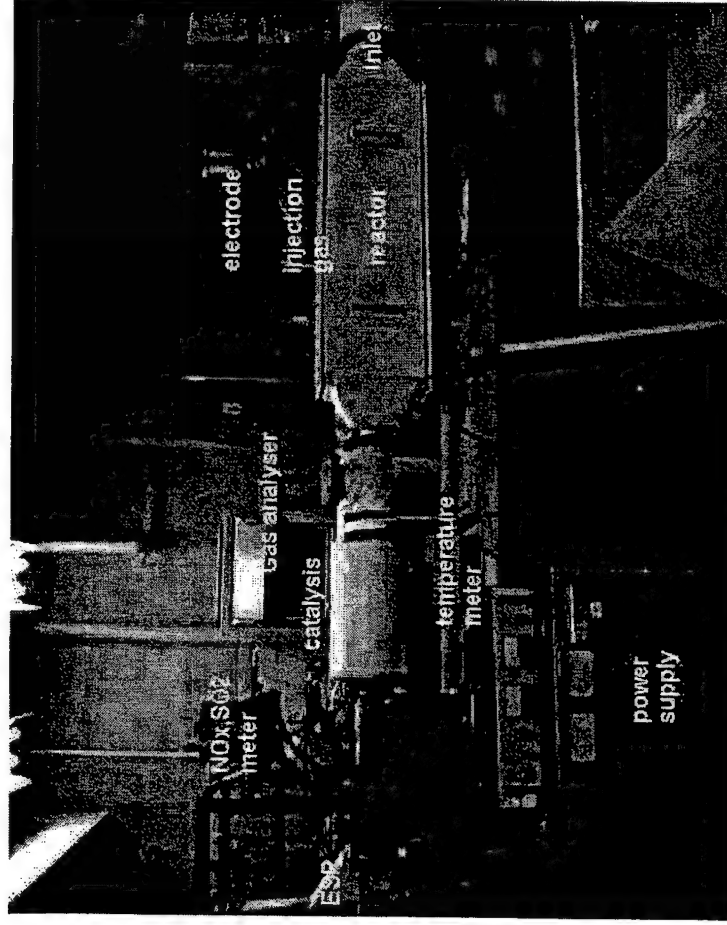
We have provided economic analyses for three hybrid NTP systems compared to conventional de-NO$_x$ systems

Conventional	NTP
Selective Catalytic Reduction (SCR) + Wet Scrubbers	Pulsed Corona
Selective Catalytic Reduction (SCR) + Electrostatic Precipitator (ESP)	Electron-Beam
	Corona Radical Shower (CRS)

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Example: NO_x -removal power requirement depends on exhaust gas flow rate, characteristic specific energy, and desired degree of removal.

(Assume stand-alone NTP system.)

- **Typical removal scaling:** $[X]/[X]_0 = \exp(-E_s/\beta)$,

where $E_s = P/Q$, plasma power divided by gas flow rate and β = specific energy for one e-fold removal.

For NO, $\beta \sim 10$ J/lit (e-beam); ~ 50 J/lit (electrical discharge).

- **Power requirement for one e-fold removal:** $P = Q \cdot \beta$.

Small Source:		Large Source:	
$Q \sim 100$ SCFM (2832 lit/min)		$Q \sim 1 \times 10^6$ SCFM (2.83×10^7 lit/min)	
$P \sim 472$ W (e-b), 2.4 kW (e-d)		$P \sim 4.7$ MW (e-b), 24 MW (e-d)	

Benchmarking Basis/Examples for Economic Analyses

Reactor Type:		Pulsed Corona [1]	Corona Shower [2]	Electron Beam [3]	Conventional [4]
Facility:		JETC Exhaust	'JETC' Exhaust	Auto Tunnel	Power Plant
Parameter	Units	Values			
$[NO]_0$	ppm	36	40	10	300
NO_x Removal	%	56	90	70	80
$[SO2]_0$	ppm	4	4	1	3,050
SO_x Removal	%	90	95	95	90
Energy Yield	g-NO/kW-h	20	17	19	-
Exp. Scale	Nm^3/h	600	12	40,000	1.9×10^6

1) Haythornthwaite et al 1997, 2) Matsuoka et al 1997 (simulated JETC exhaust), 3) Ebara Co. 1998, 4) EPRI 1983, JMIA-EEI 1991.

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Cost comparisons for various de-NO$_x$ systems (70% de-NO$_x$, 108 h/wk operation)
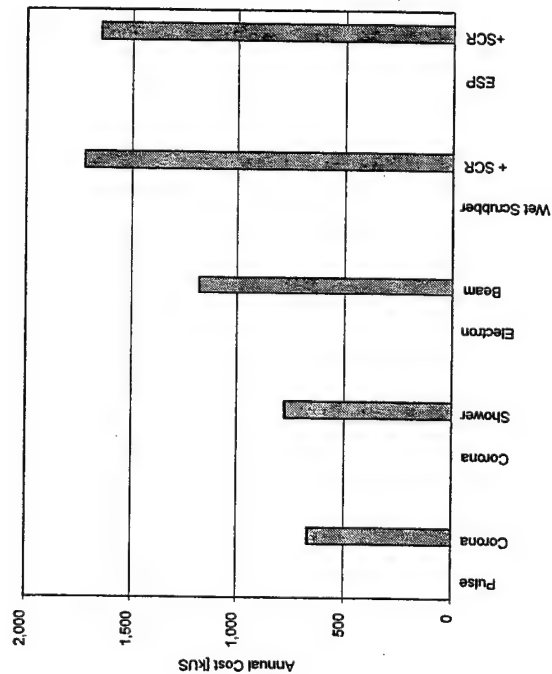
Gas Flow Rate: 1.0 x 10^5 Nm³/h (5.89 x 10^4 SCFM)

(k$)	Pulsed Corona	Corona Shower	Electron Beam	Wet Scrubber + SCR	ESP + SCR
Capital Recovery	345	410	718	36	104
Labor & Maintenance	218	246	377	273	273
Electric Power	99	116	77	123	83
Chemicals & Utilities	6	6	6	1,291	1,192
Total Annual Cost	664	774	1,176	1,723	1,651
Fertilizer Recovery	4	4	4	0	0

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Cost comparisons for various de-NO$_x$ systems (cont'd.) (70% de-NOx, 108 h/wk operation)

Annual Cost [kUS$]
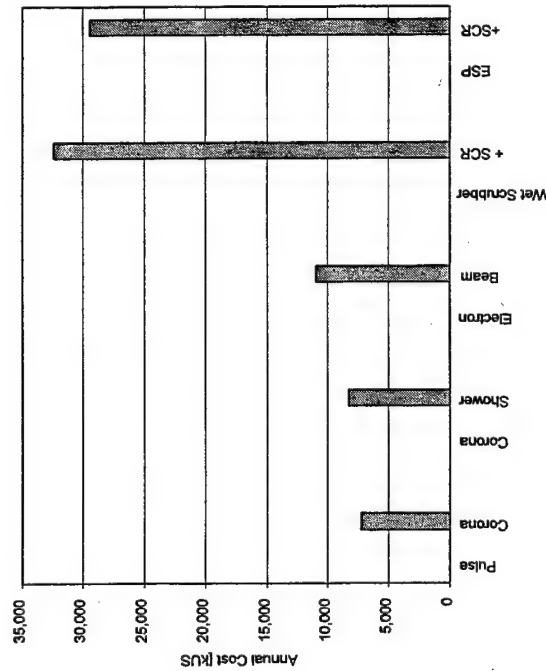
Gas Flow Rate: 1.70 x 10⁶ Nm³/h (1.0 x 10⁶ SCFM)

(k$)	Pulsed Corona	Corona Shower	Electron Beam	Wet Scrubber + SCR	ESP + SCR
Capital Recovery	3,594	4,151	6,504	3,802	3,123
Labor & Maintenance	1,823	2,060	3,061	4,641	4,641
Electric Power	1,674	1,970	1,318	2,082	1,403
Chemicals & Utilities	110	110	110	21,935	20,247
Total Annual Cost	7,139	8,230	10,931	32,459	29,414
Fertilizer Recovery	62	62	62	0	0

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Other workers have evaluated two additional NTP de-NO_x & de-SO_x/NO_x systems

Haythornthwaite et al
(Air Force contract)

(Pulsed corona oxidizer + scrubber)

Tecogen/Thermo Power
(Modified dielectric barrier)

$NO \rightarrow NO_2$ Conversion + Scrubber System

NO + VOCs + Air → **NTP Reactor** → NO_2 HNO_3 → **Wet Scrubber** → Dilute HNO_3 (w / $Na_2S_2O_3$)
NaNO$_3$ + H_2O (w / NaOH)
Ca(NO_3)$_2$ + H_2O (w / Ca(OH)$_2$)
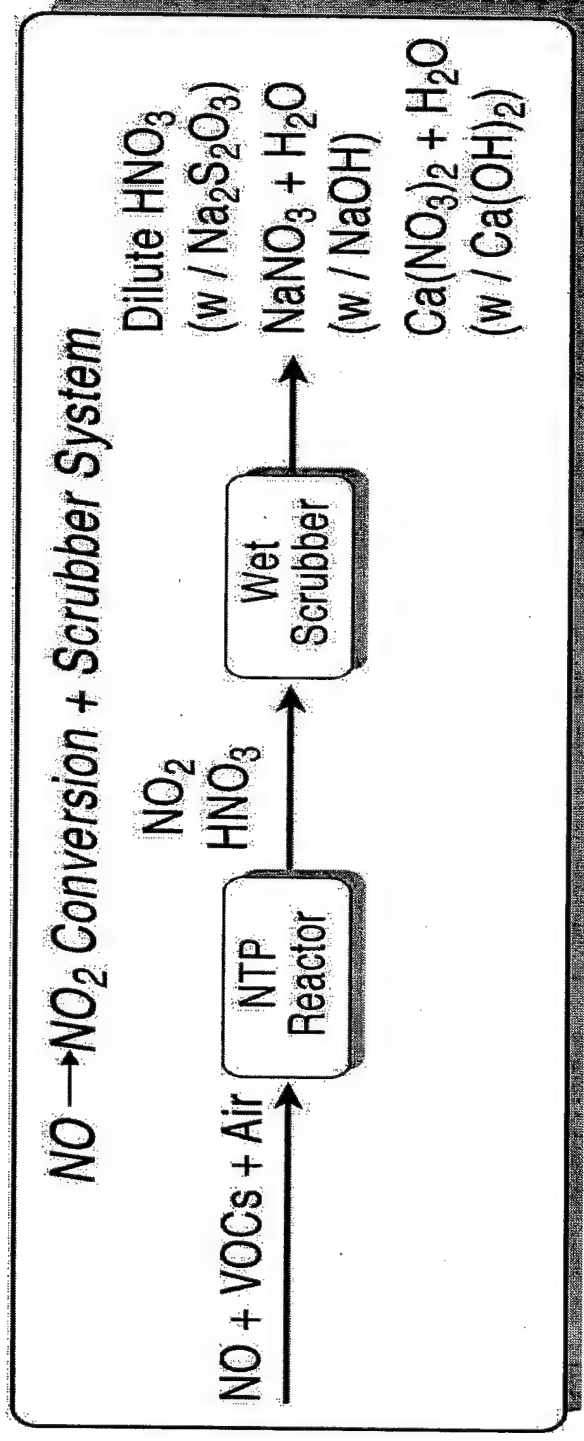
Cost data for
Haythornthwaite et al 1997
4×10^6 SCFM JETC using PCR
$NO \rightarrow NO_2$ converter + wet scrubber
($\sim$50% NO_x removal)

Capital Cost	$109,200,000	
Capital Recovery	$14,196,000	

Run Time	10 hr/week	50 hr/week
O & M Costs/Yr	$523,600	$2,618,000
Annual Cost	$14,719,600	$16,814,000

—— Los Alamos ——

Cost comparisons for commercial Tecolytic™ modified dielectrc-barrier NTP reactor system for de-NO$_x$/SO$_x$ (flue-gas treatment) show favorable trends

500 MW Power Plant

Tecolytic Technology™
(Modified dielectric barrier)

SCR: NO$_x$ w/ PM.5 and HAPs control

SCR: NO$_x$ only

Tecolytic Technology™ : NO$_x$, PM2.5 and HAPs for 2% of plant power

NO$_x$ Removed (%)

NO$_x$ Removal Cost ($/ton)
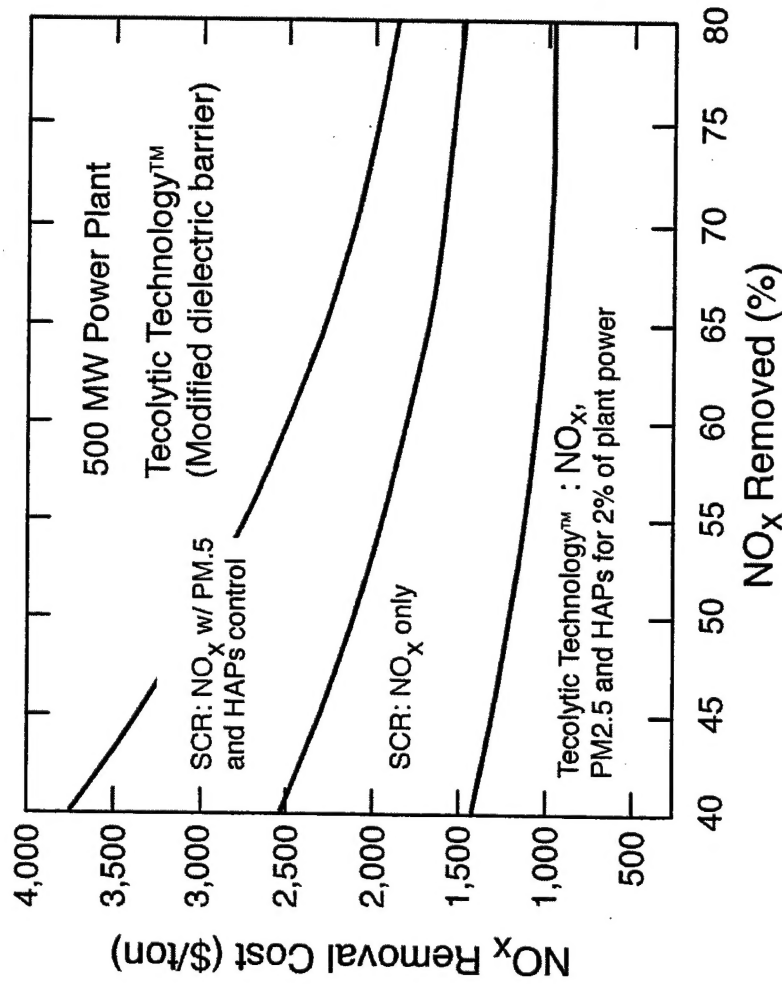
Future trends

Realizing the performance and economic shortcomings of stand-alone NTP reactors, the use of staged or hybrid systems to better match particular air-emissions control applications is being more widely explored

- NTP + injectants
- NTP + catalysts
- NTP + adsorbents

NTP technology may become more common in areas where it has significant advantages over conventional technologies in terms of:

- Increased efficiency and/or economics
- Increased selectivity or throughput
- Better control of final products and/or process
- Unique process streams

——————— Los Alamos ———————

Summary

- A greater awareness of the effects of air pollutants (NO_x/SO_x, VOCs, particulates) on our environment and human health has led to more stringent regulations on air emissions.

- Non-thermal plasma technology is being explored as an emissions-control option in several areas (this talk has emphasized NO_x control in jet engine test facilities).

- Hybrid systems (e.g., using injectants) show increased removal yields and favorable economic trends for large-scale de-NO_x systems.

- Rigorous pilot-plant tests are required to provide further data and operating experience to more fully evaluate economic and performance projections.

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